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(REV 11-98)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

## TRANSMITTAL LETTER TO THE UNITED STATES

INMAN-00714

DESIGNATED/ELECTED OFFICE (DO/EO/US)

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR

CONCERNING A FILING UNDER 35 U.S.C. 371

09/890157

INTERNATIONAL APPLICATION NO.

INTERNATIONAL FILING DATE

PRIORITY DATE CLAIMED

PCT/GB00/00714

28 February 2000

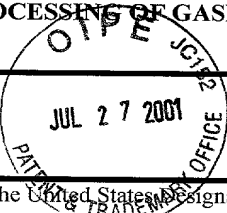
2 March 1999

TITLE OF INVENTION

PLASMA-ASSISTED PROCESSING OF GASEOUS MEDIA

APPLICANT(S) FOR DO/EO/US

INMAN, Michael



Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
  - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☒ has been transmitted by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☐ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☐ A copy of the International Search Report (PCT/ISA/210).
8. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
  - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☐ have been transmitted by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☐ have not been made and will not be made.
9. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
10. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
11. ☐ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
12. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).

## Items 13 to 20 below concern document(s) or information included:

13. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☒ A **FIRST** preliminary amendment.
16. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
17. ☐ A substitute specification.
18. ☐ A change of power of attorney and/or address letter.
19. ☐ Certificate of Mailing by Express Mail
20. ☐ Other items or information:

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.492(a)(1) - (5)) : <b>09/890157</b>		INTERNATIONAL APPLICATION NO. <b>PCT/GB00/00714</b>		ATTORNEY'S DOCKET NUMBER <b>INMAN-00714</b>	
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21. The following fees are submitted:

BASIC NATIONAL FEE ( 37 CFR 1.492 (a) (1) - (5)) :				CALCULATIONS PTO USE ONLY	
<input type="checkbox"/>	Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO .....	\$1,000.00			
<input checked="" type="checkbox"/>	International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO .....	\$860.00			
<input type="checkbox"/>	International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO .....	\$710.00			
<input type="checkbox"/>	International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) .....	\$690.00			
<input type="checkbox"/>	International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) .....	\$100.00			
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				<b>\$860.00</b>	
Surcharge of <b>\$130.00</b> for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492 (e)). <input type="checkbox"/> 20 <input type="checkbox"/> 30				<b>\$0.00</b>	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	20 - 20 =	0	x \$18.00	<b>\$0.00</b>	
Independent claims	1 - 3 =	0	x \$80.00	<b>\$0.00</b>	
Multiple Dependent Claims (check if applicable). <input type="checkbox"/>				<b>\$0.00</b>	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				<b>\$860.00</b>	
Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) (check if applicable). <input type="checkbox"/>				<b>\$0.00</b>	
<b>SUBTOTAL =</b>				<b>\$860.00</b>	
Processing fee of <b>\$130.00</b> for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492 (f)). <input type="checkbox"/> 20 <input type="checkbox"/> 30			+	<b>\$0.00</b>	
<b>TOTAL NATIONAL FEE =</b>				<b>\$860.00</b>	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable). <input checked="" type="checkbox"/>				<b>\$40.00</b>	
<b>TOTAL FEES ENCLOSED =</b>				<b>\$900.00</b>	
				Amount to be: refunded	\$
				charged	\$

☐ A check in the amount of \_\_\_\_\_ to cover the above fees is enclosed.

☒ **Authorized to charge to credit card.** Credit card payment form attached.

☐ Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \_\_\_\_\_ to cover the above fees.

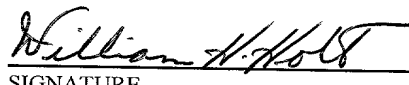
A duplicate copy of this sheet is enclosed.

☐ The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. \_\_\_\_\_ A duplicate copy of this sheet is enclosed.

**NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.**

SEND ALL CORRESPONDENCE TO:

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SIGNATURE

**William H. Holt**  
NAME

**20766**  
REGISTRATION NUMBER

**7-27-01**  
DATE

JC18 Rec'd PCT/PTO 27 JUL 2001

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of \*  
INMAN, Michael \*  
New U.S. National Stage Application \*  
of International Application No. \* Attention: DO/EO/US  
PCT/GB00/00714 \*  
International Filing Date: \*  
28 February 2000 \*  
For: PLASMA-ASSISTED PROCESSING OF GASEOUS MEDIA

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents  
Washington, D.C. 20231

Sir:

Please amend as follows:

In the Claims:

Cancel all claims presently on file, and substitute the following new Claims 21-40:

21. A reactor for the plasma assisted processing of a gaseous medium including a pair of electrodes having facing surfaces, the separation of the facing surfaces being substantially uniform and defining a space therebetween, a body of dielectric material positioned to provide a dielectric barrier between the electrodes and configured to divide the said space between the electrodes into a plurality of gas passages, which together provide the plasma volume of the reactor and along the lengths of which gas flows in use of the reactor, the gas passages being aligned so that their lengths extend between and in a direction parallel with the facing surfaces of the

electrodes, the gas passages being spaced apart from one another in a direction transverse to the said facing surfaces, wherein the gas passages are shaped to have a pair of opposed sides the contour of which matches the contour of the said facing surfaces of the electrodes, this shape and the spacing of the gas passages providing a substantially uniform distribution of electric field occurs across the plasma volume space between the electrodes.

22. A reactor according to claim 21, wherein the electrodes are embedded in a body of dielectric material which extends across the space between the electrodes and includes a plurality of gas passages extending longitudinally of the body of dielectric material to provide a plurality of electrically equivalent plasma volumes extending in series across the space between the electrodes.

23. A reactor according to claim 21, wherein the dielectric material is selected from the group consisting of alpha or gamma aluminas, cordierite, mullite, alumino silicate ceramics, silicon carbide, micaceous glass or mixtures of these.

24. A reactor according to claim 21 wherein the gas passages have surfaces which present a catalytically active surface to gaseous medium passing through them.

25. A reactor according to claim 24, wherein the said surfaces of the gas passages are coated, impregnated or treated by ion-exchange or doping, with a catalytically-active material.

26. A reactor according to claim 24, wherein the cat-

alytically active surface is catalytically active towards the reduction of the emissions of one or more of nitrogenous oxides, particulate including carbonaceous particulate, hydrocarbons including polyaromatic hydrocarbons, carbon monoxide and other regulated or unregulated combustion products from the exhausts of internal combustion engines.

27. A reactor according to claim 24, wherein the catalytically-active material is selected from the group consisting of alpha and gamma aluminas and mixtures of these phases, ferroelectric materials including of titanates, including barium titanate, titania, including anatase phase titania, zirconia, vanadia, silver aluminate, perovskites including layered perovskites and  $\text{La}_2\text{CuO}_4$ ,  $\text{La}_{1.9}\text{K}_{0.1}\text{Cu}_{0.95}\text{V}_{0.05}\text{O}_4$  and  $\text{La}_{0.9}\text{K}_{0.1}\text{CoO}_3$ , spinels, metal-doped and metal oxide-doped or exchanges inorganic oxides including cobalt oxide-doped or exchanged inorganic oxides including cobalt oxide-doped alumina, vanadates including potassium metavanadate, caesium metavanadate, pyrovanadates including potassium pyrovanadate and caesium pyrovanadate, metal-doped zeolites including zeolites known as ZSM-5, Y, beta, mordenite all of which zeolites may contain iron, cobalt or copper with or without additional catalyst promoting cations including cerium and lanthanum, alkali metal containing zeolites in particular sodium-Y zeolites and mixtures of any of these materials.

28. A reactor according to claim 21, wherein the gas passages contain a gas permeable body of an insulating filling material.

29. A reactor according to claim 28, wherein the insulating filling material comprises a dielectric material.

30. A reactor according to claim 29, wherein the dielectric material is a catalytically active material.

31. A reactor according to claim 29, wherein the dielectric material is coated, impregnated or otherwise treated with a catalytically active material.

32. A reactor according to claim 29, wherein the dielectric material develops catalytically active properties by virtue of exposure to plasma in the gas passages.

33. A reactor according to claim 21, wherein the electrodes are planar and the gas passages have a generally rectangular cross section with their major transverse dimensions parallel to those of the said facing surfaces of the electrodes.

34. A reactor according to claim 21, wherein the electrodes are in the form of two concentric cylinders and the gas passages comprise a plurality of regularly spaced slots of cylindrical form.

35. A reactor according to claim 21, wherein the arrangement of gas passages is such that the potential drop across the plasma volume between the electrodes is equal to approximately half the voltage applied to the reactor.

36. A reactor according to claim 21, wherein power supply for the reactor is provided by an integrated starter alternator damper system.

37. A reactor according to claim 21, wherein electrical-generating power supply for the reactor is provided at least

partially from the group comprising fuel cells, gas turbines, solar cells and heat exchangers.

38. A reactor according to claim 21 incorporated as part of an emissions control system.

39. A reactor according to claim 38, wherein the emissions control system is used in conjunction with an engine management system.

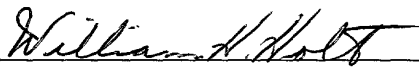
40. A reactor according to claim 38, wherein the emissions control system includes an additional gas passage outside of the plasma region of the reactor in series with the aforesaid gas passages, said additional gas passage containing gas permeable catalytically active material.

REMARKS

By this Preliminary Amendment, new claims have been added for consideration on the merits..

Favorable action is courteously solicited.

Respectfully submitted,

  
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July 27, 2001

Plasma-Assisted Processing of Gaseous Media

The present invention relates to the plasma-assisted processing of gaseous media and in particular to the  
5 reduction of the emissions of one or more of nitrogenous oxides, particulate including carbonaceous particulate, hydrocarbons including polyaromatic hydrocarbons, carbon monoxide and other regulated or unregulated combustion  
10 products from the exhausts of internal combustion engines.

One of the major problems associated with the development and use of internal combustion engines is the  
15 noxious exhaust emissions from such engines. Two of the most deleterious materials, particularly in the case of diesel engines, are particulate matter (primarily carbon) and oxides of Nitrogen ( $\text{NO}_x$ ). Increasingly severe  
20 emission control regulations are forcing internal combustion engine and vehicle manufacturers to find more efficient ways of removing these materials in particular from internal combustion engine exhaust emissions.  
Unfortunately, in practice, it is found that combustion  
25 modification techniques which improve the situation in relation to one of the above components of internal combustion engine exhaust emissions tend to worsen the  
situation in relation to the other. A variety of systems for trapping particulate emissions from internal  
30 combustion engine exhausts have been investigated, particularly in relation to making such particulate emission traps capable of being regenerated when they  
have become saturated with particulate material.

Examples of such diesel exhaust particulate filters are to be found in European patent application EP 0 010

384; US patents 4,505,107; 4,485,622; 4,427,418; and  
4,276,066; EP 0 244 061; EP 0 112 634 and EP 0 132 166.

In all the above cases, the particulate matter is  
5 removed from diesel exhaust gases by a simple physical  
trapping of particulate matter in the interstices of a  
porous, usually ceramic, filter body, which is then  
regenerated by heating the filter body to a temperature  
at which the trapped diesel exhaust particulates are  
10 burnt off. In most cases the filter body is monolithic,  
although EP 0 010 384 does mention the use of ceramic  
beads, wire meshes or metal screens as well. US patent  
4,427,418 discloses the use of ceramic coated wire or  
ceramic fibres.

15 In a broader context, the precipitation of charged,  
particulate matter by electrostatic forces also is known.  
However, in this case, precipitation normally takes place  
upon large planar electrodes of metal screens.

20 GB patent 2,274,412 discloses a method and apparatus  
for removing particulate and other pollutants from  
internal combustion engine exhaust gases, in which the  
exhaust gases are passed through a bed of charged pellets  
25 of material, preferably ferroelectric, having high  
dielectric constant. In addition to removing  
particulates by oxidation, especially electric discharge  
assisted oxidation, there is disclosed the reduction of  
NO<sub>x</sub> gases to nitrogen, by the use of pellets adapted to  
30 catalyse the NO<sub>x</sub> reduction.

Also, US patents 3 983 021, 5 147 516 and 5 284 556  
disclose the catalytic reduction of nitrogen oxides.  
However, US 3 983 021 is solely concerned with the  
35 reduction of NO to N in a silent glow discharge, the

ART 34 AMBT

- 3 -

temperature of which is kept below a value at which the oxidation of N or NO to higher oxides of nitrogen does not occur. There is no mention of any simultaneous removal of hydrocarbons.

5

US patent 5 284 556 discloses the removal of hydrocarbons from internal combustion engine exhaust emissions. The process involved is one of dissociation in an electrical discharge of the so-called 'silent' type, that is to say, a discharge which occurs between two electrodes at least one of which is insulated. The device described is an open discharge chamber. Mention is made of the possible deposition of a NO<sub>x</sub>-reducing catalyst on one of the electrodes.

15

A conventional dielectric barrier plasma assisted gas reactor such as that disclosed in specification US 5,284,556, consists of a plasma volume situated between two electrodes at least one of which has a dielectric barrier in the form of a thick layer of an insulating medium on its inner surface. JP-A-4027414 also discloses a dielectric barrier type of reactor in which electrodes, arranged parallel to the direction of gas flow, are positioned on opposite sides of a dielectric material through which bored holes provide gas passages.

In order to generate a plasma in such a device, the potential within the space between the electrodes must reach a critical value before the plasma will ignite. The potential which appears across the main plasma volume is dependent upon the ratio of the capacitance of the dielectric layer and that of the plasma volume because these two entities create a capacitive potential divider. The potential across the plasma volume is inversely proportioned to its capacitance, that is to say, the

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- 4 -

higher the capacitance of the plasma volume, the lower is the potential difference across it. This effect can cause a serious problem if the plasma volume is filled with a gas permeable material which has a high dielectric constant, such as pellets of barium titanate, because the potential difference across such a reactor bed may never reach the critical value for the plasma to ignite unless the supply voltage is very high, of the order of tens of kilovolts which may exceed the safe working voltage of the dielectric barrier, or other high voltage components of the power supply.

It is an object of the present invention to provide an improved reactor for the plasma-assisted processing of a gaseous medium.

According to the invention in one aspect there is provided a reactor for the plasma assisted processing of a gaseous medium including a pair of electrodes having facing surfaces, the separation of the facing surfaces being substantially uniform and defining a space therebetween, and a body of dielectric material positioned to provide a dielectric barrier between the electrodes (1,2;21,22) and configured to divide the said space between the electrodes (1,2;21,22) into a plurality of gas passages (6;24), which together provide the plasma volume of the reactor and along the lengths of which gas flows in use of the reactor, the gas passages (6;24) being aligned so that their lengths extend between and in a direction parallel with the facing surfaces of the electrodes (1,2;21,22), the gas passages being spaced apart from one another in a direction transverse to the said facing surfaces, characterised in that the gas passages (6;24) are shaped so as to have a pair of opposed sides the contour of which matches the contour of the said facing surfaces of the electrodes (1,2;21,22),

- 5 -

this shape and the spacing of the gas passages being such that a substantially uniform distribution of electric field occurs across the plasma volume space between the electrodes.

5

According to the present invention in a second aspect there is provided a reactor wherein the electrodes are embedded in a body of dielectric material which extends across the space between the electrodes and

10 includes a plurality of gas passages extending longitudinally of the body of dielectric material to provide a plurality of electrically equivalent plasma volumes extending in series across the space between the electrodes.

15

Preferably the matrix of gas passages in the dielectric medium between the electrodes is adapted to provide a potential difference across the plasma volume space between the electrodes equal to half the supply

20 voltage. The potential difference across this space will vary according to gas flow, temperature and gas composition and therefore the overall size and shape and position of location of the reactor within the exhaust system is selected so as to ensure that variances from

25 this optimum condition are minimized.

The surfaces of the gas passages can be coated, impregnated or generally treated by for example ion exchange or doping with a material which is catalytically

30 active in relation to the gas and or particulate processing reactions to be carried out in the reactor, or the gas passages can be filled with a gas permeable form of such a material. The geometry of the gas passages or the gas permeable filling material, which may be

35 dielectric, can then be adapted to negate the capacitive effects of the catalytic material. Alternatively, the

- 6 -

dielectric material of the reactor can itself be chosen to be catalytic in relation to the gas and or particulate processing reactions to be carried out in the reactor. The gas permeable material filling the passages can be

5 catalytically-active or non-catalytically thermally-active with respect to processing of the gaseous medium although a catalytically-active material may be present on the surface of non-catalytic dielectric material contained in the reactor as a coating or it may be

10 present on or in the gas permeable filling material by generally treating the material by for example ion exchange or doping.

Gas permeable dielectric filling material for the

15 reactor can be in the form of spheres, pellets, extrudates, fibres, sheets, wafers, frits, meshes, coils, foams, membrane, ceramic honeycomb monolith or granules or as a coating on any of the above shapes or on a ceramic foam or ceramic honeycomb monolith. In addition

20 to optimising the plasma discharge and gas processing characteristics, combinations of one or more of the above can be used to create a filter structure with a non-uniform surface area and porosity, for example a graded porosity when presented to the exhaust gas particularly

25 when containing particulates as described in patent specification WO00/43102. Gas permeable dielectric filling material that can be placed inside the reactor can also be housed outside of the plasma region of the reactor so that the gaseous media can either pass through

30 this material before entering the plasma region or pass through this material after passing through the plasma region. When placed outside the reactor, dielectric filling material can be replaced by ceramic, polymeric or metallic material in the same form described above for

35 the dielectric filling material. Dielectric filling material can act as a selective filter as described in

- 7 -

the specification of our PCT/GB00/3943 . respectively.  
Dielectric material or trapped species on it in the  
plasma can be made to appear to act as a catalytic  
surface to the gas processing reactions even though  
5 neither the plasma nor the dielectric material nor  
trapped species alone need have catalytic properties as  
described in the specification of our application  
PCT/GB00/3943.

10 When the gaseous medium comprises the exhaust gases  
containing nitrogenous oxides and particulate material  
such as carbonaceous particulate that are derived from an  
internal combustion engine supplied with combustion fuel,  
the exhaust gases can contain hydrocarbon either added  
15 separately or residually derived from the fuel  
combustion. The exhaust can contain a chemical additive  
acting as a carbon combustion catalyst that is either  
present initially in the fuel or added separately to the  
exhaust and whose function is to lower the combustion  
20 temperature and/or increase the rate of removal of  
carbonaceous material. Carbon combustion catalyst can be  
encapsulated within or bound to a fugitive additive that  
chemically decomposes during or shortly after fuel  
combustion thus releasing the additive into the fuel or  
25 exhaust. Examples of carbon combustion catalysts are  
alkali-metal salts such as lithium nitrate described in  
GB 2 232 613 B, cerium oxide, alkali-metal doped  
lanthanum oxide-vanadium oxide, perovskites such as  
 $\text{La}_{0.9}\text{K}_{0.1}\text{CoO}_3$  and also layered perovskites or vanadate or  
30 combinations of such materials although such carbon  
combustion catalysts can also constitute all or part of  
the dielectric filling material described above. The  
mode of operation of such catalysts is described in our  
specification W000/43102. The use of a carbon combustion  
35 catalyst can reduce the power requirements to the plasma

- 8 -

reactor for treating carbonaceous particulate material and reduce the volume of active material.

For the reduction of nitrogenous material for which  
5 zeolites are particularly useful materials, the plasma  
can produce activated hydrocarbon from hydrocarbon  
reductant in the exhaust as described in our publication  
WO99/12638 and/or convert nitrogenous oxides to nitrogen  
dioxide as described in WO99/12638 and WO00/43102. It  
10 should be appreciated that material that is not catalytic  
for the reduction of nitrogenous material when not  
exposed to a plasma may develop catalytic properties for  
this reduction when exposed to a plasma due for example  
to activation by O atoms or other plasma-generated free  
15 radicals or activation by plasma generated species such  
as activated hydrocarbons and or nitrogen dioxide. It  
should be appreciated that the dielectric filling  
material can also be placed outside the plasma zone and  
outside the reactor with a multiplicity of additive  
20 injection ports as described in WO99/12638. Catalytic  
properties can be further augmented by the electric field  
and or other charged species present in or adjacent to  
the plasma region. A reductant other than hydrocarbon  
may be used, in particular nitrogen containing species  
25 such as ammonia, urea or cyanuric acid. When a nitrogen  
containing species is used as a reductant for  
nitrogenous oxide reduction a particularly useful  
catalyst is vanadium pentoxide-titanium dioxide. When  
using a nitrogen containing reductant species, mixing  
30 with effluent can also be made after the effluent has  
passed through the plasma zone of the reactor before  
contact with the catalyst.

The invention will now be described, by way of  
35 example, with reference to the accompanying drawings, in  
which

- 8a -

Figure 1 is a perspective view of the operative part of a reactor embodying the invention for the plasma assisted processing of a gaseous medium, and

- 5        Figure 2 is a transverse section of a second embodiment of the invention.

Referring to Figure 1 of the drawings, the operative part of a reactor for the plasma assisted processing of a  
10        gaseous medium includes two planar electrodes 1 and 2 to one of which is connected a high voltage supply cable 3. The other electrode has a cable 4 connected to it by

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means of which it can be connected to a suitable grounding point. The electrodes 1 and 2 are surrounded and separated by a body 5 of a dielectric material, which supports the electrodes so that there is a substantially uniform separation of their facing surfaces. The dielectric material is required to maintain thermal stability across a range of exhaust gas temperatures, and is selected from such materials as alpha and gamma aluminas, aluminosilicate ceramics, cordierite, silicon carbide, mullite or a moldable dielectric ceramic material such as the micaceous glass MICATHERM as disclosed in our patent specification WO99/20373. A number of identical gas passages 6 extend through the body 5 of dielectric material parallel to the electrodes 1 and 2 and spaced apart from one another in a direction transverse to the said facing surfaces so that a uniform distribution of electric field occurs across the space between the electrodes.

The gas passages 6 are separated by regions 7 of dielectric medium which are of equal thickness and parallel to the electrodes 1 and 2 so as to form a distributed dielectric barrier between the electrodes 1 and 2.

The electrical isolation of the gas passages 6 from one another prevents the plasma from forming a short circuit between the electrodes 1 and 2. Also, it can be shown that the maximum coupling of power into the plasma volume between the electrodes 1 and 2 occurs when the potential drop across the plasma volume is equal to half the supply voltage. This can be achieved by a suitable choice of the number and cross-section of the gas channels 6. The supply voltage can be derived from a resonant power supply that is adjacent thus in close proximity to the reactor as described in our publications

- 10 -

WO99/05400 and the specification of our application  
WO00/43645.

5 If desired, the gas channels 6 can be filled with a  
gas permeable body made of an insulating material which  
is catalytic or non-catalytic towards the gas and or  
particulate processing reactions to be carried out in the  
reactor. Alternatively, the surfaces of the gas passages  
6 or gas permeable filling material can be coated with  
10 such a catalytic material, or the entire body of  
dielectric can be made of such a material. The choice of  
material, which can by itself be catalytic or non-  
catalytic in the presence or absence of the plasma,  
depends on the requirements to process nitrogeneous  
15 oxides or particulate material and other emissions  
described previously. Dielectric material or trapped  
species on it in the plasma can be made to appear to act  
as a catalytic surface to the gas processing reactions  
even though neither the plasma nor the dielectric  
20 material nor trapped species alone need have catalytic  
properties. Gas permeable dielectric filling material  
that can be placed inside the reactor can also be housed  
outside of the plasma region of the reactor so that the  
gaseous media can either pass through this material  
25 before entering the plasma region or passes through this  
material after passing through the plasma region. It will  
be appreciated that the same material can be used in the  
plasma zone as outside the plasma zone or combinations of  
different materials can be used in the plasma zone and  
30 outside the plasma zone and that exhaust gas and or  
particulate processing reactions can be carried out by  
combinations of identical or different materials in or  
out of the plasma zone.

35 In practice, of course, the operative part of the  
reactor is contained in an envelope which includes inlet

- 11 -

and outlet stubs by means of which it can be incorporated in pipework through which the gaseous medium to be processed is caused to flow and means for ensuring that all the said gaseous medium passes through the gas passages 6.

Although the invention has been described in terms of a planar geometry as shown in figure 1, it is equally applicable to a cylindrical geometry as shown in Figure 2, although in this case, the radial thicknesses of the gas passages will have to vary in order that a uniform radial potential drop be achieved. The embodiments of reactor described in these examples may include catalytic components or be installed as part of an emissions control system employing catalysts or other emission control devices for the plasma assisted treatment of the exhaust gases from internal combustion engines. Such other emission control devices may comprise exhaust gas recirculation (EGR), variations in ignition timing, fuel injection timing and fuel injection pulse rate shaping. The reactor of these examples can be used in conjunction with a power supply and engine management system as described in patent specification W000/50746. An article 'Stop go systems get the green light' in European Automotive Design, April 1998, pages 24-26 describes an example of an integrated starter alternator damper system (ISAD). Such an ISAD can be used as part of a power supply system to power a plasma assisted emissions control system of which a reactor as described herein is part. In addition, other power sources such as but not limited to fuel cells, gas turbines, solar cells and heat exchangers can be the primary or part-provider of the electrical-generating power source that can also be used to power the power supply system for the reactor.

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Referring to Figure 2 a cylindrical reactor body 20 consists of two cylindrical electrodes 21 and 22, which are embedded co-axially in a cylindrical body 23 of a temperature resistant dielectric material. Between the electrodes 21 and 22 are a series of concentric gas passages 24 of cylindrical form separated by radial supporting webs 25. The gas passages 24 are so arranged that they are superimposed radially and each group of gas passages 24 corresponds to the series of gas passages 6 of the Figure 1 embodiment of the invention. (Only two rings of gas passages are shown in the drawing although it will be appreciated that the invention is not restricted to use of only two of such rings).

Unlike the Figure 1 embodiment of the invention, the radial widths of the gas passages 24 are not the same, but are a function of the distance of the centres of the gas passages 24 from the inner electrode 22. This is because the radial distribution of the electric field between the electrodes 21 and 22 also is a function of the radial distance from the surface of the inner electrode 22. By a suitable choice of radial widths for the gas passages 24, these two effects can be made to cancel out, giving substantially equal radial electric fields in each of the gas passages 24.

In this case, the inner electrode 22 is arranged to be the high voltage electrode.

A particular use for such reactors is to reduce the emissions of one or more of nitrogeneous oxides, particulate including carbonaceous particulate, hydrocarbons including polyaromatic hydrocarbons, carbon monoxide and other regulated or unregulated combustion products from the exhausts of internal combustion engines. In this case, suitable dielectric materials for

- 13 -

the body 5, are alpha and gamma aluminas, cordierite, mullite, aluminosilicate ceramics, silicon carbide, micaceous moldable ceramics such as MICATHERM or mixtures of these. Suitable catalytic material that can be used

5 for coating the surfaces of the gas channels 6, or for use as the dielectric filling material or for depositing onto the dielectric filling material, for example as a coating, are aluminas known under their Registered Trade Marks as LD 350, CT 530, Condea hollow extrudates, DYPAC,

10 T-60 Alumina, T-162 alumina cordierite,  $\alpha$ ,  $\chi$  and  $\gamma$  aluminas, and aluminas containing mixtures of these phases, ferroelectric materials such as titanates particularly barium titanate; titania, particularly in the anatase phase; zirconia, vanadia, silver aluminate,

15 perovskites, spinels, metal-doped and metal oxide-doped or exchanged inorganic oxides such as cobalt oxide-doped alumina, vanadates and pyrovanadates and metal-doped zeolites. Examples of zeolites are those known as ZSM-5, Y, beta, mordenite all of which may contain iron, cobalt

20 or copper with or without additional catalyst promoting cations such as cerium and lanthanum. Other examples of zeolites are alkali metal containing zeolites in particular sodium-Y zeolites that are particularly useful for treatment of nitrogenous oxides. Examples of

25 perovskites are  $\text{La}_2\text{CuO}_4$ ,  $\text{La}_{1.9}\text{K}_{0.1}\text{Cu}_{0.95}\text{V}_{0.05}\text{O}_4$  and  $\text{La}_{0.9}\text{K}_{0.1}\text{CoO}_3$ . Examples of vanadates are potassium metavanadate, caesium metavanadate, potassium pyrovanadate and caesium pyrovanadate. Mixtures of these compounds can also be used.

30

Gas permeable dielectric filling material for the reactor can be in the form of spheres, pellets, extrudates, fibres, sheets, wafers, frits, meshes, coils, foams, membrane, ceramic honeycomb monolith or granules

- 13a -

or as a coating on a ceramic foam or ceramic honeycomb monolith. Combinations of one or more of the above can be

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used to create a filter structure with a non-uniform surface area and porosity, for example a graded porosity when presented to the exhaust gas and the filling material can also be placed outside of the plasma region  
5 of the reactor.

Zeolite materials are particularly useful for treatment of nitrogeneous oxides while perovskites and vanadates are particularly useful for treatment of  
10 carbonaceous particulates. Perovskites can also be particularly useful for combined removal of nitrogeneous oxides and particulate material.

Thus, exhaust gases containing hydrocarbon reductant  
15 residually-derived from the fuel combustion or added separately or containing a nitrogen-containing reductant for reduction of nitrogeneous oxides and or containing a carbon combustion catalyst as described earlier is passed through the plasma region of the plasma reactor  
20 containing dielectric material that is catalytically-active for gaseous processing reactions or is passed through such material before entering the plasma region or is passed through such material after passing through the plasma region. When a nitrogen-containing reductant  
25 is used the reductant can be added to exhaust leaving the plasma region of the reactor and before passage over catalytically-active dielectric material.

Also, suitable supply voltages are obtained from a  
30 power supply adapted to produce pulses having a potential of the order of kilovolts to tens of kilovolts and repetition frequencies in the range 50 to 5000 Hz, although higher frequencies of the order of tens of kilohertz can be used. Pulsed direct current is  
35 convenient for automotive use, but alternating potentials

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for example triangular or sine waves of the same or similar characteristics can be used.

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- 16 -

Claims

1. A reactor for the plasma assisted processing of a gaseous medium including a pair of electrodes (1,2;21,22) having facing surfaces, the separation of the facing surfaces being substantially uniform and defining a space therebetween, a body (5;23) of dielectric material positioned to provide a dielectric barrier between the electrodes (1,2;21,22) and configured to divide the said space between the electrodes (1,2;21,22) into a plurality of gas passages (6;24), which together provide the plasma volume of the reactor and along the lengths of which gas flows in use of the reactor, the gas passages (6;24) being aligned so that their lengths extend between and in a direction parallel with the facing surfaces of the electrodes (1,2;21,22), the gas passages being spaced apart from one another in a direction transverse to the said facing surfaces, characterised in that the gas passages (6;24) are shaped so as to have a pair of opposed sides the contour of which matches the contour of the said facing surfaces of the electrodes (1,2;21,22), this shape and the spacing of the gas passages being such that a substantially uniform distribution of electric field occurs across the plasma volume space between the electrodes (1,2;21,22).

2. A reactor according to claim 1, further characterised in that the electrodes (1,2;21,22) are embedded in a body (5;23) of dielectric material which extends across the space between the electrodes (1,2;21,22) and includes a plurality of gas passages (6;24) extending longitudinally of the body (5;23) of dielectric material to provide a plurality of electrically equivalent plasma volumes extending in series across the space between the electrodes

- 16a -

3. A reactor according to claim 1 or claim 2, further  
characterised in that the dielectric material is selected  
from the group consisting of alpha or gamma aluminas,  
cordierite, mullite, alumino silicate ceramics, silicon  
5 carbide, micaceous glass or mixtures of these.

4. A reactor according to claim 1 or claim 2 or claim  
3, further characterised in that the gas passages (6;24)

are adapted to present a catalytically active surface to gaseous medium passing through them.

5. A reactor according to claim 4, further  
5 characterised in that the surfaces of the gas passages (6;24) are coated, impregnated or treated by ion-exchange or doping, with a catalytically-active material.
6. A reactor according to claim 4 or claim 5, further  
10 characterised in that that the catalytically active surface is catalytically active towards the reduction of the emissions of one or more of nitrogenous oxides, particulate including carbonaceous particulate, hydrocarbons including polyaromatic hydrocarbons, carbon  
15 monoxide and other regulated or unregulated combustion products from the exhausts of internal combustion engines.
7. A reactor according to claim 4 or claim 5 or claim  
20 6, further characterised in that the catalytically-active material is selected from the group comprising alpha and gamma aluminas and mixtures of these phases, ferroelectric materials such as titanates, including barium titanate, titania, including anatase phase  
25 titania, zirconia, vanadia, silver aluminate, perovskites including layered perovskites and  $\text{La}_2\text{CuO}_4$ ,  $\text{La}_{1.9}\text{K}_{0.1}\text{Cu}_{0.95}\text{V}_{0.05}\text{O}_4$  and  $\text{La}_{0.9}\text{K}_{0.1}\text{CoO}_3$ , spinels, metal-doped and metal oxide-doped or exchanged inorganic oxides including cobalt oxide-doped alumina, vanadates including  
30 potassium metavanadate, caesium metavanadate, pyrovanadates including potassium pyrovanadate and caesium pyrovanadate, metal-doped zeolites including zeolites known as ZSM-5, Y, beta, mordenite all of which zeolites may contain iron, cobalt or copper with or  
35 without additional catalyst promoting cations such as

cerium and lanthanum, alkali metal containing zeolites in particular sodium-Y zeolites and mixtures of any of these materials.

- 5 8. A reactor according to any of the preceding claims, further characterised in that the gas passages(6;24) contain a gas permeable body of an insulating filling material.
- 10 9. A reactor according to claim 8, further characterised in that the insulating filling material comprises a dielectric material.
- 15 10. A reactor according to claim 9, further characterised in that the dielectric material is a catalytically active material.
- 20 11. A reactor according to claim 9 or claim 10, further characterised in that the dielectric material is coated, impregnated or otherwise treated with a catalytically active material.
- 25 12. A reactor according to claim 9, further characterised in that the dielectric material develops catalytically active properties by virtue of exposure to plasma in the gas passages(6;24).
- 30 13. A reactor according to any of the preceding claims, further characterised in that the electrodes(1,2;21,22) are planar and the gas passages(6;24) have a generally rectangular cross-section with their major transverse dimensions parallel to those of the said facing surfaces of the electrodes(1,2;21,22).
- 35 14. A reactor according to any of claims 1 to 12, further characterised in that the electrodes(21,22) are

- 19 -

in the form of two concentric cylinders and the gas passages(24) comprise a plurality of regularly spaced slots of cylindrical form.

- 5 15. A reactor according to any of the preceding claims, further characterised in that the arrangement of gas passages (6,24) is such that the potential drop across the plasma volume between the electrodes is equal to approximately half the voltage applied to the reactor.
- 10 16. A reactor according to any of the preceding claims, further characterised in that power supply for the reactor is provided by an integrated starter alternator damper system.
- 15 17. A reactor according to any of claims 1 to 15, further characterised in that fuel cells, gas turbines, solar cells or heat exchangers are used as primary or part-provider of an electrical-generating power supply
- 20 for the reactor.
18. A reactor according to any of the preceding claims incorporated as part of an emissions control system.
- 25 19. A reactor according to claim 18, further characterised in that the emissions control system is used in conjunction with an engine management system.
- 30 20. A reactor according to claim 18 or claim 19, further characterised in that the emissions control system includes an additional gas passage outside of the plasma region of the reactor in series with the aforesaid gas passages(6,24), said additional gas passage containing gas permeable catalytically active material.
- 35

21. A reactor according to claim 20, further characterised in that the catalytically active material in the said additional gas passage comprises a dielectric material.
- 5
22. A reactor according to claim 20, further characterised in that the catalytically active material in the said additional gas passage comprises a polymeric or metallic material.
- 10
23. A reactor according to any of claims 20, 21 or 22, further characterised in that the gas permeable catalytically active material in the said additional gas passage is in the form of spheres, pellets, extrudates, 15 fibres, sheets, wafers, frits, meshes, coils, foams, membrane, ceramic honeycomb monolith or granules or as a coating on a ceramic foam or ceramic honeycomb monolith.
- 20
24. A method of plasma assisted processing of a gaseous medium using a reactor according to any of the preceding claims, characterised in that a reductant for the treatment of nitrogeneous oxides is supplied to the said gas passages(6,24) or said additional gas passage.
- 25
25. A method according to claim 24, further characterised in that the reductant comprises a hydrocarbon or a nitrogen containing compound.
- 30
26. A method according to claim 25, further characterised in that the reductant comprises hydrocarbon residually derived from fuel combustion.
- 35
27. A method according to claim 25, further characterised in that the reductant comprises a nitrogen containing compound selected from ammonia, urea or cyanuric acid.

Fig.1.

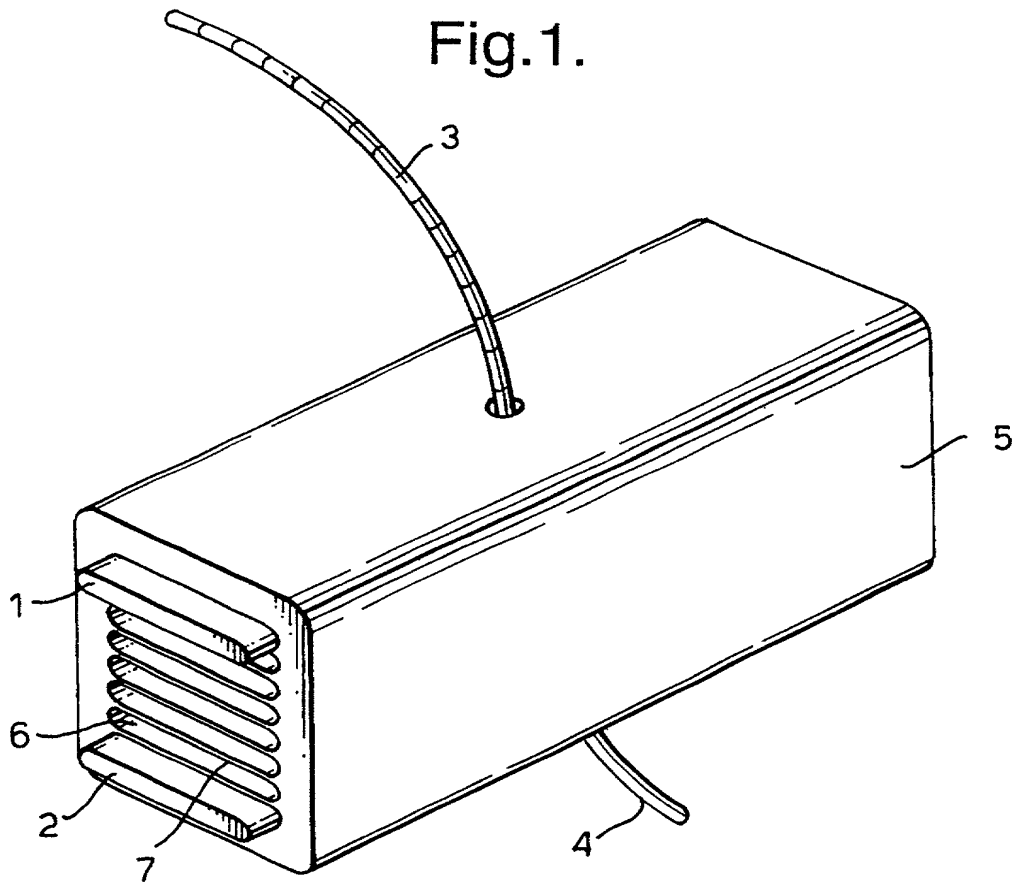
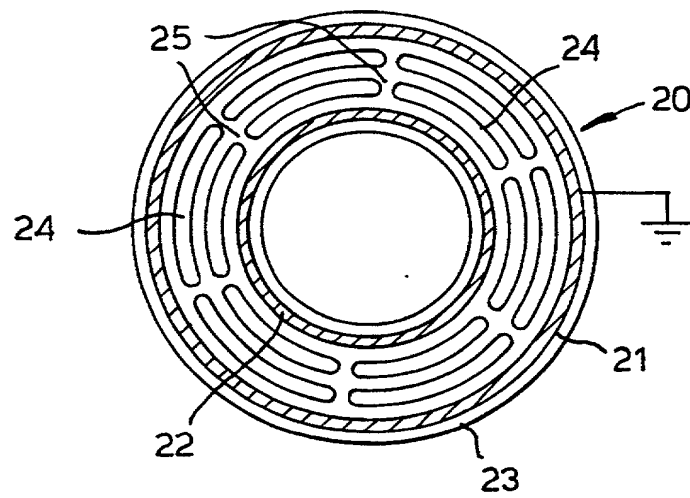


Fig.2.



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**COMBINED DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION (USA)**  
(INCLUDING DESIGN PATENT APPLICATIONS)

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

**Plasma assisted processing of gaseous media**

the specification of which is attached hereto, unless the following box is checked:

☐ was filed on 28<sup>th</sup> February 2000 as United States Application Number or PCT International Application No. PCT/GB00/00714 and was amended on 24<sup>th</sup> February 2001 (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

<u>99 04640.1</u> (Number)	<u>United Kingdom</u> (Country)	<u>28<sup>th</sup> February 1999</u> (Day/Month/Year Filed)	<table border="0"><tr><td>Priority Claimed</td></tr><tr><td><input checked="" type="checkbox"/> Yes <input type="checkbox"/> No</td></tr></table>	Priority Claimed	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
Priority Claimed					
<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No					
<u>                    </u> (Number)	<u>                    </u> (Country)	<u>                    </u> (Day/Month/Year Filed)	<table border="0"><tr><td>Priority Claimed</td></tr><tr><td><input type="checkbox"/> Yes <input type="checkbox"/> No</td></tr></table>	Priority Claimed	<input type="checkbox"/> Yes <input type="checkbox"/> No
Priority Claimed					
<input type="checkbox"/> Yes <input type="checkbox"/> No					

☐ Additional applications identified on attached sheet.

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

<u>                    </u> (Application Serial Number)	<u>                    </u> (Filing Date)	<u>                    </u> (Status) (patented, pending, abandoned)
<u>                    </u> (Application Serial Number)	<u>                    </u> (Filing Date)	<u>                    </u> (Status) (patented, pending, abandoned)

☐ Additional applications identified on attached sheet.

I hereby appoint the following attorneys to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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☐ See attached sheet for similar information and signatures for additional joint inventors.

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